

Magnetic behavior of van Vleck ions and an electron gas
interacting by exchange

by

L. Palermo
Instituto de Física da UFF
Niterói, R.J., Brasil

X.A. da Silva
Centro Brasileiro de Pesquisas Físicas
Rio de Janeiro, R.J., Brasil

ABSTRACT

The magnetic behavior of a model in which van Vleck ions, under the action of a crystal field, interacting by exchange with an electron gas is investigated. The condition of onset of ferromagnetism and the behavior of the critical temperature, band and ionic magnetizations (and susceptibilities) versus temperature, as a function of the band width, exchange interaction and the crystal field splitting energy parameters are obtained within an approximation equivalent to a molecular field formulation.

On a étudié le comportement magnétique d'un modèle dans lequel des ions van Vleck, sous l'action d'un champ cristallin, interagissent par échange avec un gaz d'électrons. On a obtenu la condition d'existence d'ordre ferromagnétique, la dépendance avec la température de l'aimantation et de la susceptibilité ainsi que la température critique en fonction de la largeur de bande et des paramètres du champ cristallin et d'échange dans une approximation de champ moléculaire.

1. INTRODUCTION

The onset of ferromagnetism in a system consisting of a paramagnetic electron gas and localized magnetic moments coupled in such way that the electron gas creates a molecular field which acts on the localized spins and vice versa is, since long, an exercise in text books [1]. This model, sometimes referred to as Vonsoviskii-Zener (V-Z) model, has been extensively used as the starting point to explain a variety of magnetic properties (magnetic susceptibilities, Curie temperatures, phase transitions) of rare earth intermetallic compounds [2-5]. In the case of the heavy rare earth compounds of Y and Co a quantitative prediction of the Curie temperature as a function of the rare earth position in the periodic table can be successfully compared to the experimental data [6]. However, for the light rare earth compounds, as remarked by Bloch et al [5], crystal field effects have to be taken into account and that complicates the simple description applicable to the heavy rare earth compounds.

A particularly interesting case where the crystal field plays a fundamental role is the case of the systems where the rare earth component is a van Vleck ion. A typical example is PrAl_2 for which magnetization and susceptibility studies as a function of temperature are available [7,8]. These are tentatively explained with the help of a model, known as bootstrap ferromagnetic model [9], in which in addition to a crystal field term (describing the two level splitting of the ground state of the van Vleck ion), a Heisenberg interaction between the pseudo-spins associated to the ions. In a

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molecular field approximation this model exhibits ferromagnetism. Here, in contrast to the V-Z model, the conduction band does not seem to play a major role although one may assume it is somehow implicit in the exchange interaction.

The purpose of this work is to explore the magnetic properties of the following model: a crystal of van Vleck ions coexists with a paramagnetic electron gas. Each ion is under the action of a crystal field, which splits the ground state into two levels and interacts, via exchange, with the itinerant electrons. We show that this model may exhibit ferromagnetism both in the electron gas and in the ions, the magnetizations and susceptibilities being a function of the band, crystal field and exchange parameters. The plan of the paper is as follows. In the next section we introduce the model, the physical quantities to be computed and the method employed. In section 3 the formal solution and the approximations used are presented. Finally, in section 4, the numerical solutions and the magnetic behavior are discussed.

2. FORMULATION OF THE PROBLEM

The model Hamiltonian is

$$H = \sum_{i,j,\sigma} T_{ij} c_{i\sigma}^+ c_{j\sigma} - \Delta \sum_i S_i^X - J \sum_i S_i^Z s_i^Z \quad (1)$$

The first term is the conduction band Hamiltonian in the Wannier representation. In the Bloch representation it is written as

$$\sum_{k\sigma} \epsilon_k c_{k\sigma}^+ c_{k\sigma}$$

where

$$T_{ij} = \frac{1}{N} \sum_{\vec{k}} \epsilon_{\vec{k}} e^{-i\vec{k} \cdot (\vec{R}_i - \vec{R}_j)}$$

and

$$c_{\vec{k}\sigma}^{\dagger} = \frac{1}{\sqrt{N}} \sum_i e^{i\vec{k} \cdot \vec{R}_i} c_{i\sigma} \quad (c_{\vec{k}\sigma}^{\dagger} = \frac{1}{\sqrt{N}} \sum_i e^{-i\vec{k} \cdot \vec{R}_i} c_{i\sigma}^{\dagger})$$

\vec{k} and \vec{R}_i are defined in the reciprocal and direct space respectively; the sum over \vec{k} is in the Brillouin Zone and $\sigma = \uparrow$ or \downarrow .

The second and third terms represent the ions Hamiltonian and the exchange interaction between the conduction electrons and the electrons of the ions. They are an extension - taking into account symmetry translation - of a model introduced by Fulde and Peschel [10], who concentrated on the study of the dynamical susceptibility of a single van Vleck ion interacting with an electron gas. In a previous paper [11], on the electron gas spin polarization features of the Fulde-Peschel model, we also rederived the last two terms of (1) (restricted to a single ion) emphasizing the conceptual distinction between the pseudo spin S^Z and the electronic spin s^Z . For simplicity we assume that the Crystal field splits the energy level of the ions into only two levels, which are not degenerated. So S^X and S^Z are 2×2 Pauli matrices and Δ is the energy difference between the sub-levels.

The effective J is given by:

$$J = 4J'(g_i - 1)\alpha$$

where α is the matrix element of the angular momentum between the two levels of the ions, g_i the Lande's factor and J' is the exchange integral of the localized and itinerant electrons.

Our aim is to study the band and ion magnetizations. These quantities are respectively

$$m = g_e \mu_B \langle s^Z \rangle \quad (2-a)$$

$$M = 2(g_i \alpha) \mu_B \langle S^Z \rangle \quad (2-b)$$

where g_e is the gyromagnetic factor of the conduction electron and μ_B the Bohr magneton.

$\langle S^Z \rangle$ and $\langle s^Z \rangle$ are the statistical averages associated to the ion pseudo-spin and conduction electron spin polarization. In order to compute these objects we start, similarly to a procedure used previously [11], from the relations

$$S_i^Z = \frac{1}{2} - S_i^- S_i^+$$

$$s_i^Z = \frac{1}{2} (c_{i\uparrow}^+ c_{i\uparrow} - c_{i\downarrow}^+ c_{i\downarrow}) \quad (3-b)$$

Due to symmetry translation $\langle S_i^Z \rangle$ and $\langle s_i^Z \rangle$ should be independent of i and we can drop sub indices in the statistical averages. Furthermore we also have

$$\langle s^Z \rangle = \frac{1}{2N} \sum_k \langle (c_{k\uparrow}^+ c_{k\uparrow} - c_{k\downarrow}^+ c_{k\downarrow}) \rangle \quad (3-c)$$

The statistical averages $\langle S^- S^+ \rangle$ and $\langle c_{k\sigma}^+ c_{k\sigma} \rangle$ ($\sigma = \uparrow$ or \downarrow) are to be obtained using the double time Green function method.

Once these are available (see section 3) the following questions, as a function of the parameters of the model, are considered

- a) the condition of the onset of magnetic order and the behavior of the saturation magnetizations at $T = 0$.
- b) The Curie temperature.
- c) The temperature dependence of the magnetizations and the susceptibilities.

These subjects are treated in the last section.

3. APPROXIMATIONS AND FORMAL SOLUTION

In order to compute $\langle s^z \rangle$ and $\langle S^z \rangle$, using equations 3, we define the Green functions $\langle c_{i\sigma} | c_{j\sigma}^+ \rangle$ and $\langle S_i^+ | S_i^- \rangle$. Following the same steps of [11], but now having in mind the translation symmetry, we easily see that $\langle c_{i\sigma} | c_{j\sigma}^+ \rangle$ and $\langle S_i^+ | S_i^- \rangle$ depend on $\langle S_i^z c_{i\sigma} | c_{j\sigma}^+ \rangle$ and $\langle s_i^z S_i^+ | S_i^- \rangle$ respectively.

Making the decouplings

$$\langle S_i^z c_{i\sigma} | c_{j\sigma}^+ \rangle \approx \langle S^z \rangle \langle c_{i\sigma} | c_{j\sigma}^+ \rangle \quad (4-a)$$

$$\langle s_i^z S_i^+ | S_i^- \rangle \approx \langle s^z \rangle \langle S_i^+ | S_i^- \rangle \quad (4-b)$$

we obtain

$$\langle c_{k\sigma} | c_{k'\sigma}^+ \rangle = \frac{\delta_{kk'}}{2\pi} \frac{1}{\omega - (\epsilon_k - J\sigma \langle S^z \rangle - \mu)} \quad (5)$$

where $\langle c_{k\sigma} | c_{k'\sigma}^+ \rangle$ is $\langle c_{i\sigma} | c_{j\sigma}^+ \rangle$ in its Bloch representation. In (5) μ is the chemical potential and σ as a subindice is \uparrow or \downarrow ; as a coefficient it is $+$ or $-$ respectively.

The following equations determine $\langle S_i^+ | S_i^- \rangle$

$$(\omega - J \langle S^Z \rangle) \langle S_i^+ | S_i^- \rangle = \frac{1}{2\pi} - \Delta \langle S_i^Z | S_i^- \rangle \quad (6-a)$$

$$\omega \langle S_i^Z | S_i^- \rangle = -\frac{\Delta}{2} \langle S_i^+ | S_i^- \rangle + \frac{\Delta}{2} \langle S_i^- | S_i^- \rangle \quad (6-b)$$

$$(\omega - J \langle S^Z \rangle) \langle S_i^- | S_i^- \rangle = \Delta \langle S_i^Z | S_i^- \rangle \quad (6-c)$$

Solving equations 6 for $\langle S_i^+ | S_i^- \rangle$ and using the Zubarev's algorithm [12], which connects the Green function $\langle A | B \rangle$ to the statistical average $\langle BA \rangle$, we obtain

$$\langle S^Z \rangle = \frac{J \langle S^Z \rangle}{2(J^2 \langle S^Z \rangle^2 + \Delta^2)^{1/2}} \tanh \left[\frac{(J^2 \langle S^Z \rangle^2 + \Delta^2)^{1/2}}{2kT} \right] \quad (7)$$

Similarly from (5) and (3-c)

$$\langle S^Z \rangle = \left[\frac{1}{2N} \int_0^\infty \frac{n(\epsilon) d\epsilon}{\exp \left[\frac{\epsilon - (J \langle S^Z \rangle + \mu)}{kT} \right] + 1} - \int_0^\infty \frac{n(\epsilon) d\epsilon}{\exp \left[\frac{\epsilon - (-J \langle S^Z \rangle + \mu)}{kT} \right] + 1} \right] \quad (8)$$

$$N = \int_0^\infty \frac{n(\epsilon) d\epsilon}{\exp \left[\frac{\epsilon - (J \langle S^Z \rangle + \mu)}{kT} \right] + 1} + \int_0^\infty \frac{n(\epsilon) d\epsilon}{\exp \left[\frac{\epsilon - (-J \langle S^Z \rangle + \mu)}{kT} \right] + 1} \quad (9)$$

The last equation, relates the chemical potential μ to N , the number of electrons in the band; $n(\epsilon)$ is the band density of states. Introducing the band width parameter ϵ_0 as $2 \int_0^{\epsilon_0} n(\epsilon) d\epsilon = N$ and from now on assuming a parabolic density of states $n(\epsilon) = \frac{3}{4} \frac{N}{(\epsilon_0)^{3/2}} \sqrt{\epsilon}$, we rewrite (8) and (9) as

$$\frac{4}{3} \left(\frac{\epsilon_0}{kT} \right)^{3/2} \xi = F(\eta + \beta) - F(\eta - \beta) \quad (10)$$

$$\frac{4}{3} \left(\frac{\epsilon_0}{kT} \right)^{3/2} = F(\eta + \beta) + F(\eta - \beta) \quad (11)$$

where

$$\xi = 2 \langle S^z \rangle, \beta = \frac{J \langle S^z \rangle}{kT}, \eta = \frac{\mu}{kT}$$

and $F(\eta) = \int_0^\infty \frac{x^{1/2} dx}{\exp(x - \eta) + 1}$ is the Stoner-Mc Dougall function [13].

The equations 7, 10 and 11 are our formal solution. From them we may obtain ξ (the band magnetization), $\langle S^z \rangle$ and η , in terms of the parameters $\frac{\Delta}{\epsilon_0}$ and $\frac{J}{\epsilon_0}$, as a function of kT/ϵ_0 . It is interesting to note that equations 10 and 11 go into the Stoner equations 3.5 and 3.4 of reference 13. if we make the correspondence $J \langle S^z \rangle \rightarrow k\theta \xi$, where $k\theta$ is the intraband exchange in the Stoner notation. In this way we may say that in our case the electron gas sees a molecular field induced by the ions which in its turn feel, equation (7), a molecular field proportional to $J \langle s^z \rangle$ (see Appendix) created by the electrons in a process of mutual magnetization.

4. Numerical solution and magnetic behavior

We have solved the system of equations (7), (10) and (11) with the help of the McDougall-Stoner tables for $F(\eta)$ and $F'(\eta)$, available in the range $-4 \leq \eta \leq 20$ [14]. Initially we look for the condition of the onset of ferromagnetism and the saturation magnetization of the band and ions at $T = 0$. After that we determine the Curie temperature, choosing model parameters which allow us to use the McDougall-Stoner tables. Finally, the temperature dependence of magnetizations and susceptibilities are obtained.

4.1 - Magnetization at $T = 0$

At $T = 0$, (7), (10) and (11) reduce to

$$2\langle S^z \rangle = \frac{\xi}{|\xi^2 + 4\left(\frac{\Delta}{J}\right)^2|^{1/2}} \quad (12)$$

$$(1+\xi)^{2/3} - (1-\xi)^{2/3} = \frac{J}{\epsilon_0} \frac{\xi}{|\xi^2 + 4\left(\frac{\Delta}{J}\right)^2|^{1/2}} \quad (13)$$

We have solved (12) and (13) for ξ and $2\langle S^z \rangle$. Figures 1a and 1b show the band and ion magnetizations, respectively (for increasing values of Δ/ϵ_0) as function of J/ϵ_0 . One notes in (a) and (b) that for greater values of Δ/ϵ_0 one needs greater J/ϵ_0 in order to start the magnetization process, the onset of magnetization occurring when $\frac{\Delta}{\epsilon_0} = \frac{3}{8}\left(\frac{J}{\epsilon_0}\right)^2$. The magnetizations, as a function of J/ϵ_0 , increase more slowly for higher Δ/ϵ_0 . The behavior of ξ and $2\langle S^z \rangle$ in figure 1 reflect the process of mutual magnetization as a function of the crystal field and exchange.

4.2 - Curie Temperature

We look in (7), (10) and (11) for $T = T_c$, defined as: $T \rightarrow T_c$ when ξ and $\langle S^z \rangle$ goes to zero we obtain

$$4 \left(\frac{\Delta}{J} \right) \frac{kT_c}{J} = \frac{F'(\eta_c)}{F(\eta_c)} \tanh \frac{\Delta}{2kT_c} \quad (14)$$

$$\left(\frac{kT_c}{\epsilon_0} \right)^{3/2} = \frac{2}{3} \frac{1}{F(\eta_c)} \quad (15)$$

From (14) and (15) we obtain kT_c/ϵ_0 as a function of J/ϵ_0 and Δ/ϵ_0 . Figure 2a shows kT_c/ϵ_0 as a function of J/ϵ_0 , for increasing values of Δ/ϵ_0 ; figure 2b is similar to 2a, but with different Δ/J values. One notes, in 2a, that for the same J/ϵ_0 value, kT_c/ϵ_0 increases as Δ/ϵ_0 decreases.

4.3 - Temperature dependence of the magnetizations

In this section we present the band, ionic and total magnetization as a function of temperature, using the same model parameters used in 4.1 and 4.2. Figures 3a and 3b show respectively ξ and $2\langle S^z \rangle$ versus kT/ϵ_0 for different values of J/ϵ_0 and Δ/ϵ_0 . The role of Δ/ϵ_0 on the magnetization behavior is displayed by comparing curves 1, 2 and 4 with the curve 3.

Figure 3c shows a comparison of the band magnetization between the Stoner behavior and our case. The parameters Δ/ϵ_0 and J/ϵ_0 were chosen in such a way that both curves have the same kT_c/ϵ_0 and the same magnetization at $T = 0$. One notes that the Stoner magnetization is never greater than ours.

Finally, figure 3d shows the total magnetization

$$\frac{M_T}{\mu_B} = 2g_i\alpha \langle S^Z \rangle + g_e \langle s^Z \rangle \quad (16)$$

In constructing figure 3d we have taken $g_e = 2$ and $g_i\alpha = 2$.

4.4 - Temperature dependence of the magnetic susceptibilities

Above T_c , only under the action of a magnetic field H the system exhibits a magnetization; this comes both from the band and the ions. In the limit $H \rightarrow 0$ we obtain $\mu_B H / \xi \epsilon_0$ and $\mu_B H / 2 \langle S^Z \rangle \epsilon_0$. These quantities are directly related to the inverse susceptibilities of the band and ions, respectively.

$$\frac{\mu_B H}{\xi \epsilon_0} = \frac{1 - \frac{F'(\eta)}{F(\eta)} \frac{\left(\frac{J}{\epsilon_0}\right)^2}{4 \frac{\Delta}{\epsilon_0} \frac{kT}{\epsilon_0}} \tanh \frac{\frac{\Delta}{\epsilon_0}}{2 \frac{kT}{\epsilon_0}}}{\frac{F'(\eta)}{F(\eta)} \left[\frac{1}{\frac{kT}{\epsilon_0}} + \frac{2 \frac{J}{\epsilon_0} (g_i\alpha)^2}{\frac{kT}{\epsilon_0} \frac{\Delta}{\epsilon_0}} \tanh \frac{\frac{\Delta}{\epsilon_0}}{2 \frac{kT}{\epsilon_0}} \right]} \quad (17)$$

$$\frac{\mu_B H}{2 \langle S^Z \rangle} = \frac{1 - \frac{F'(\eta)}{F(\eta)} \frac{\left(\frac{J}{\epsilon_0}\right)^2}{4 \frac{\Delta}{\epsilon_0} \frac{kT}{\epsilon_0}} \tanh \frac{\frac{\Delta}{\epsilon_0}}{2 \frac{kT}{\epsilon_0}}}{\tanh \left(\frac{\frac{\Delta}{\epsilon_0}}{2 \frac{kT}{\epsilon_0}} \right) \left[\frac{F'(\eta)}{F(\eta)} \frac{\frac{J}{\epsilon_0}}{2 \frac{kT}{\epsilon_0} \frac{\Delta}{\epsilon_0}} + \frac{8 (g_i\alpha)^2}{2 \frac{\Delta}{\epsilon_0}} \right]} \quad (18)$$

These quantities are displayed in the figures 3a and 3b (right-hand scale). The susceptibilities show Curie-Weiss behavior, except for higher values of J/ϵ_0 . Other things being

equal the susceptibilities increase with Δ/ϵ_0 . The comparison with the Stoner case and the inverse of the total susceptibility are shown in the figures 3c and 3d.

The use of the McDougall-Stoner tables for $F(\eta)$ and $F'(\eta)$ in the range $-4 \leq \eta \leq 20$ imposes restrictions on the choice of the parameters Δ/ϵ_0 and J/ϵ_0 . As a consequence the kT_c/ϵ_0 obtained are too high to allow a confrontation of our model with real systems. An extension of the McDougall-Stoner tables to higher values of η , which will eventually permit us to make a detailed comparison with the magnetization and susceptibility as a function of temperature of the PrAl_2 is in progress.

APPENDIX

The Hamiltonian of a van Vleck ion under the action of a magnetic field and a crystal field which splits the ground state into two non-degenerate levels is:

$$H = \begin{pmatrix} 0 & -\delta \\ -\delta & \Delta \end{pmatrix} \quad (1.A)$$

where Δ is the energy difference associated to the crystal field splitting and δ is the magnetic interaction, namely

$$\delta = g \alpha \mu_B h$$

g is the Lande factor, h the applied magnetic field and

$$\alpha = \langle 0 | J_z | 1 \rangle$$

Defining $S = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}$, which is unitary, and making

a similitude transformation we get

$$\tilde{H} = S^{-1}HS = -\Delta S^x - 2\delta S^z \quad (2.A)$$

where $S^x = \frac{1}{2} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$ and $S^z = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$

In order to obtain the ion magnetization one has to compute the partition function $Z = \sum_{\sigma=\pm} e^{-\beta E_{\sigma}}$, where E_{σ} is the eigenvalue defined by

$$\begin{pmatrix} -\delta & -\frac{\Delta}{2} \\ -\frac{\Delta}{2} & \delta \end{pmatrix} |\pm\rangle = E_{\pm} |\pm\rangle \quad (3.A)$$

As can be verified

$$E_{\pm} = \pm \frac{1}{2} (4\delta^2 + \Delta^2)^{1/2} \quad (4.A)$$

and

$$|\pm\rangle = \pm \frac{1}{[2(4\delta^2 + \Delta^2)]^{1/2}} \begin{pmatrix} (4\delta^2 + \Delta^2 - 4\delta E_{\pm})^{1/2} \\ (4\delta^2 + \Delta^2 + 4\delta E_{\pm})^{1/2} \end{pmatrix} \quad (5.A)$$

From Z one calculates $F = -kT \ln Z$ and finally $M = -\left(\frac{\partial F}{\partial h}\right)_T$

$$M = \frac{2g\alpha\mu_B\delta}{(4\delta^2 + \Delta^2)^{1/2}} \tanh \frac{(4\delta^2 + \Delta^2)^{1/2}}{2kT} \quad (6.A)$$

It is also interesting to calculate $\langle S^z \rangle$

$$\langle S^z \rangle = \frac{1}{Z} \sum_{\sigma=\pm} e^{-\beta E_{\sigma}} \langle \sigma | S^z | \sigma \rangle$$

Using (5) one obtains

$$\langle \pm | S^Z | \pm \rangle = \pm \frac{\delta}{(4 \delta^2 + \Delta^2)^{1/2}}$$

and

$$\langle S^Z \rangle = \frac{\delta}{(4 \delta^2 + \Delta^2)^{1/2}} \tanh \frac{(4 \delta^2 + \Delta^2)^{1/2}}{2 kT} \quad (7.A)$$

Combining (6.A) and (7.A) one gets

$$M = 2 g \alpha \mu_B \langle S^Z \rangle \quad (8.A)$$

In writing (2-b), in section 2, we are actually making the Ansatz that (8) still applies in the case where the magnetization is induced by an exchange interaction.

One finally notes that putting in (7.A)

$$\delta = \frac{J}{2} \langle S^Z \rangle$$

one recovers expression 7 of section 3.

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FIGURE CAPTIONS

- Fig. 1a - Band magnetization versus J/ϵ_0 at $T=0$. The numbers on the curves are the values of Δ/ϵ_0 .
- Fig. 1b - Ion magnetization versus J/ϵ_0 at $T=0$. The numbers on the curves are the values of Δ/ϵ_0 .
- Fig. 2a - kT_c/ϵ_0 versus J/ϵ_0 . The numbers on the curve are the values of Δ/ϵ_0 .
- Fig. 2b - kT_c/ϵ_0 versus J/ϵ_0 . The numbers on the curve are the values of Δ/J .
- Fig. 3a - Band magnetization and inverse of band susceptibility versus kT/ϵ_0 . Curves 1, 2 and 4 correspond to $\Delta/\epsilon_0 = 0.10$ and J/ϵ_0 equal to 0.73, 1.00 and 1.33 respectively. For curve 3 we have $\Delta/\epsilon_0 = 0.30$ and $J/\epsilon_0 = 1.30$. The right hand scale corresponds to the inverse of susceptibility.
- Fig. 3b - Ion magnetization and inverse of ion susceptibility. Curves 1-4 correspond to the values of Δ/ϵ_0 and J/ϵ_0 as given in figure 3.
- Fig. 3c - Band magnetization and inverse of band susceptibility. Curve 2 is a pure Stoner model. Curve 1 corresponds to our model.
- Fig. 3d - Total magnetization and inverse of total susceptibility. Curves 1-4 correspond to the values of Δ/ϵ_0 and J/ϵ_0 as given in figure 3a.

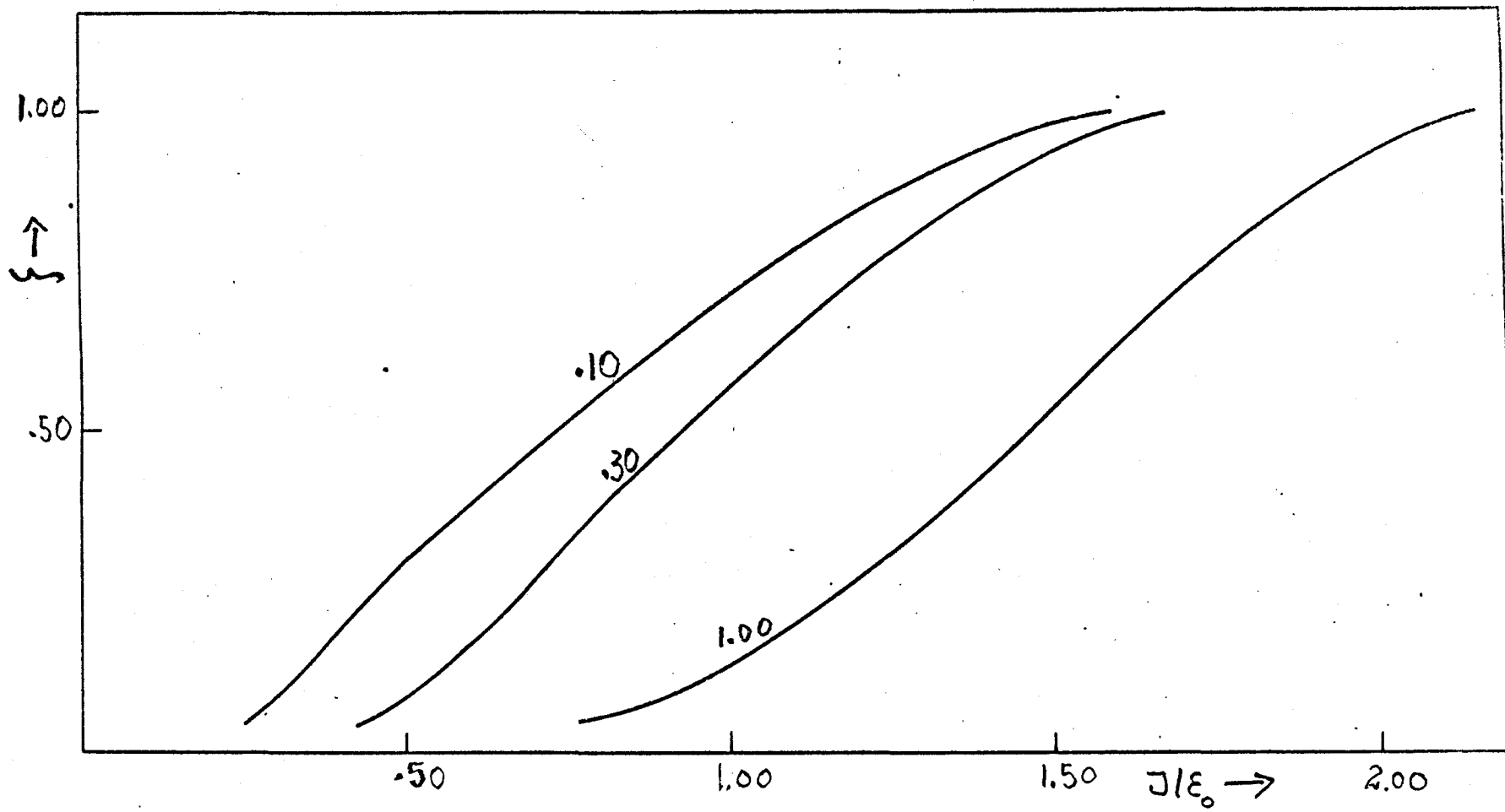


Fig. 1a

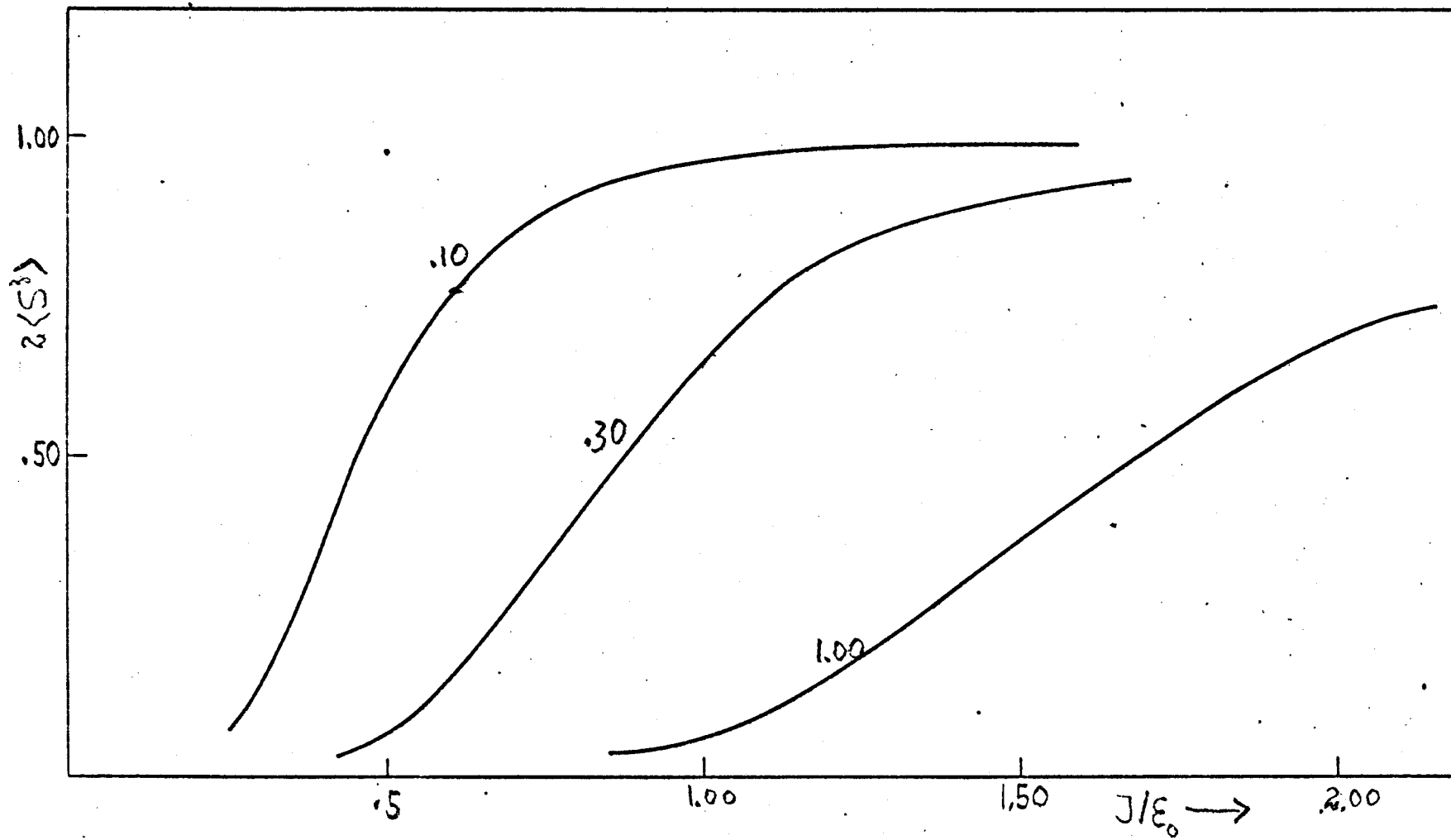


Fig. 1b

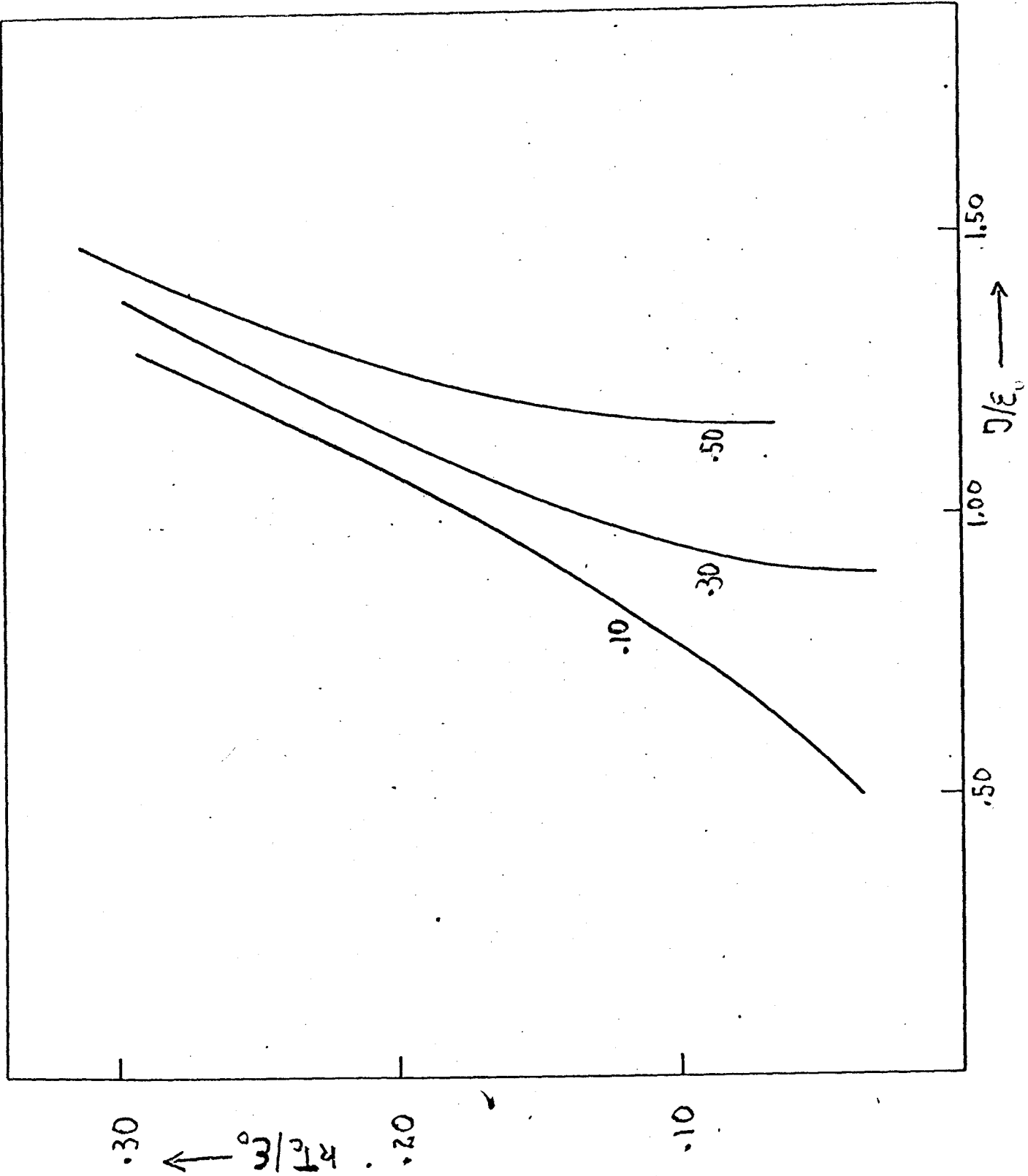


Fig. 2a

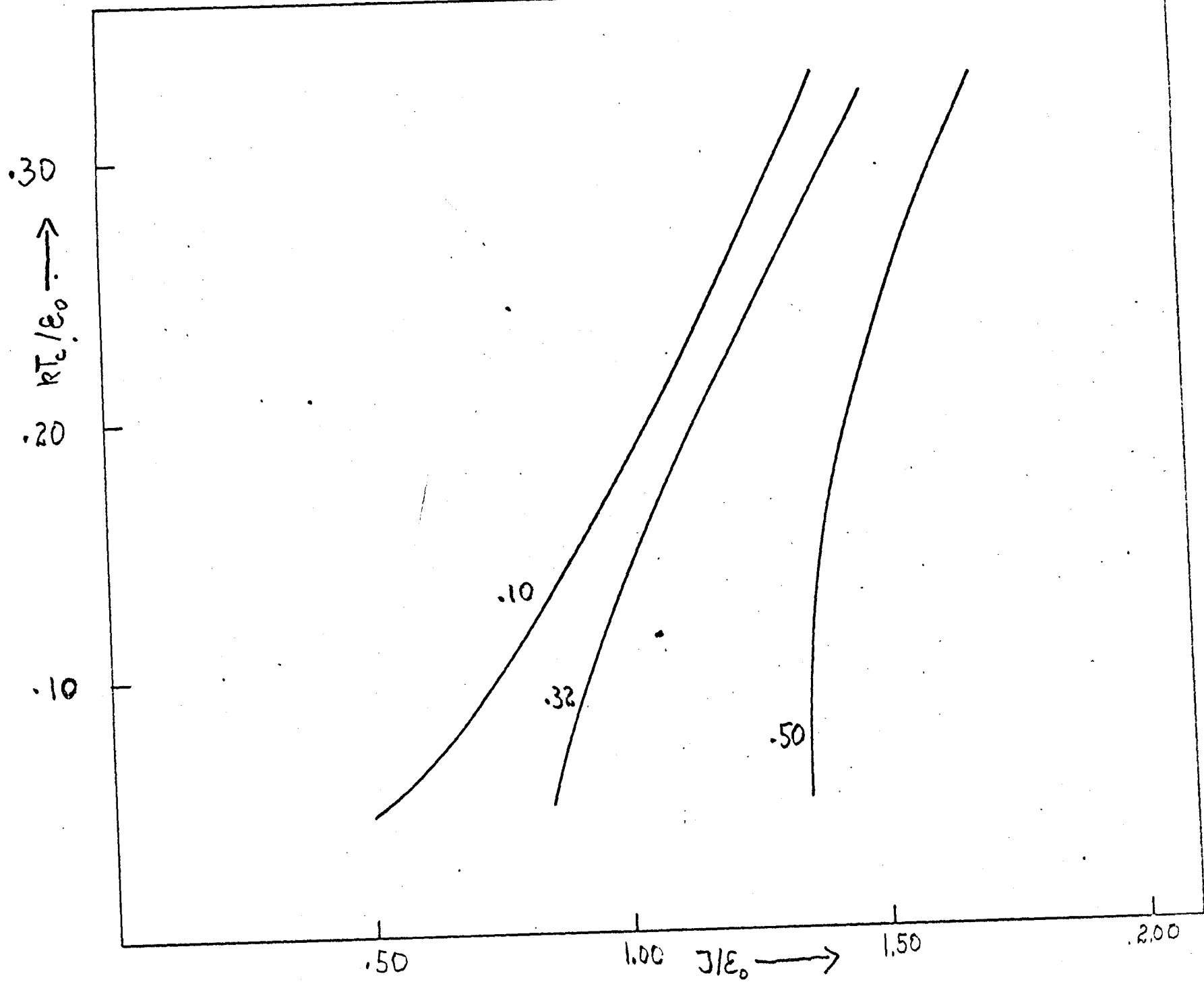


Fig. 2b

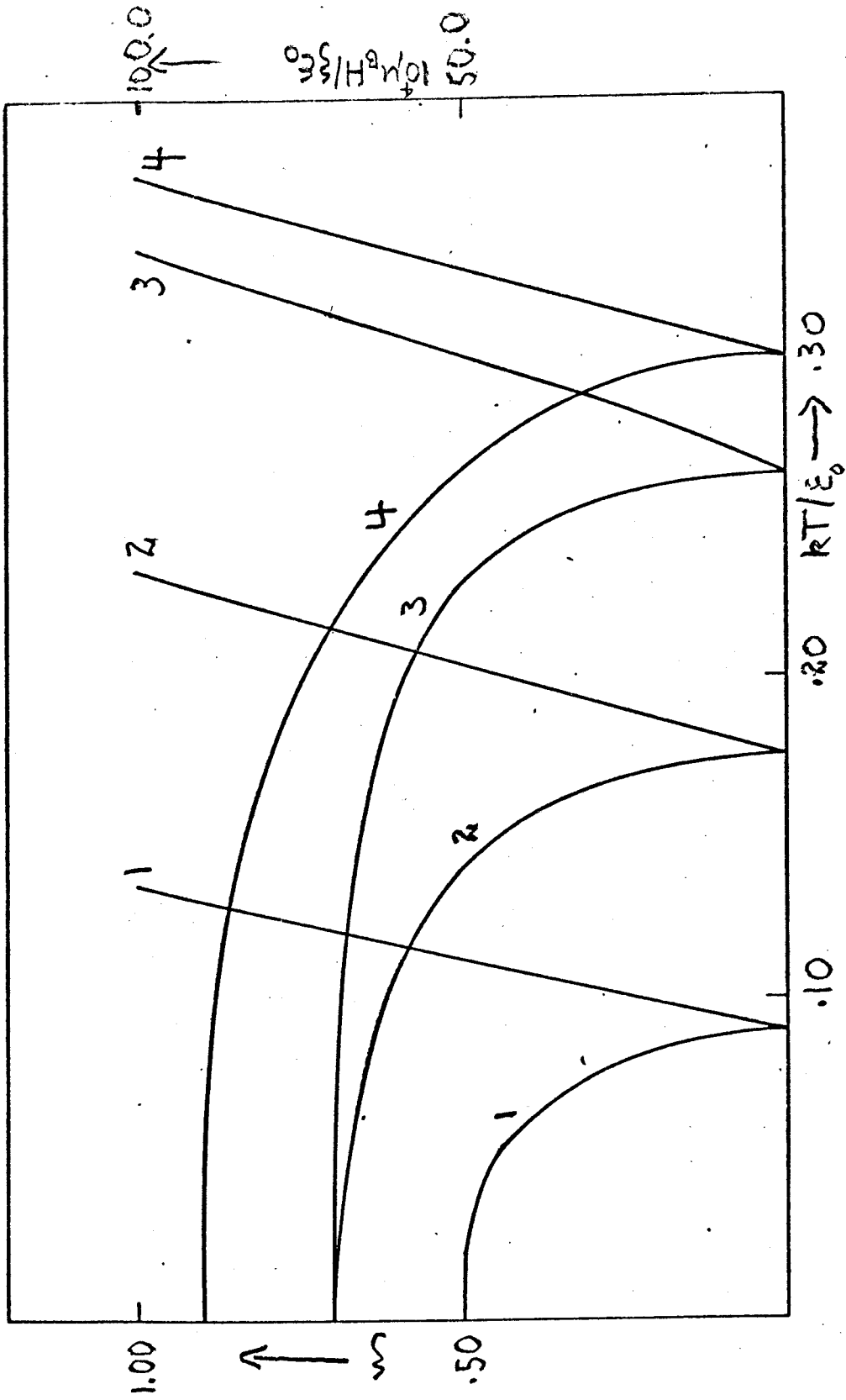


Fig. 3a

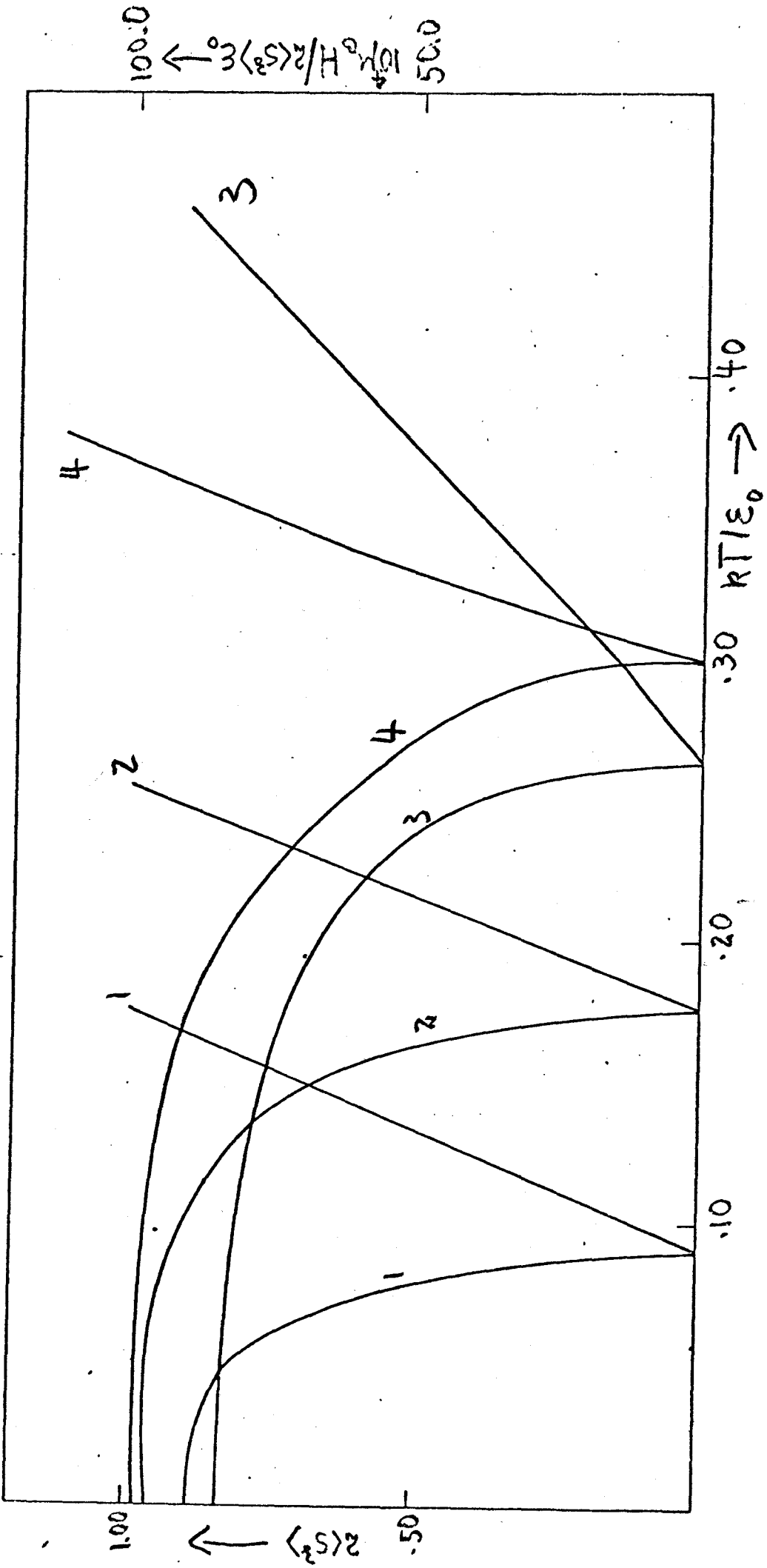


Fig. 3b

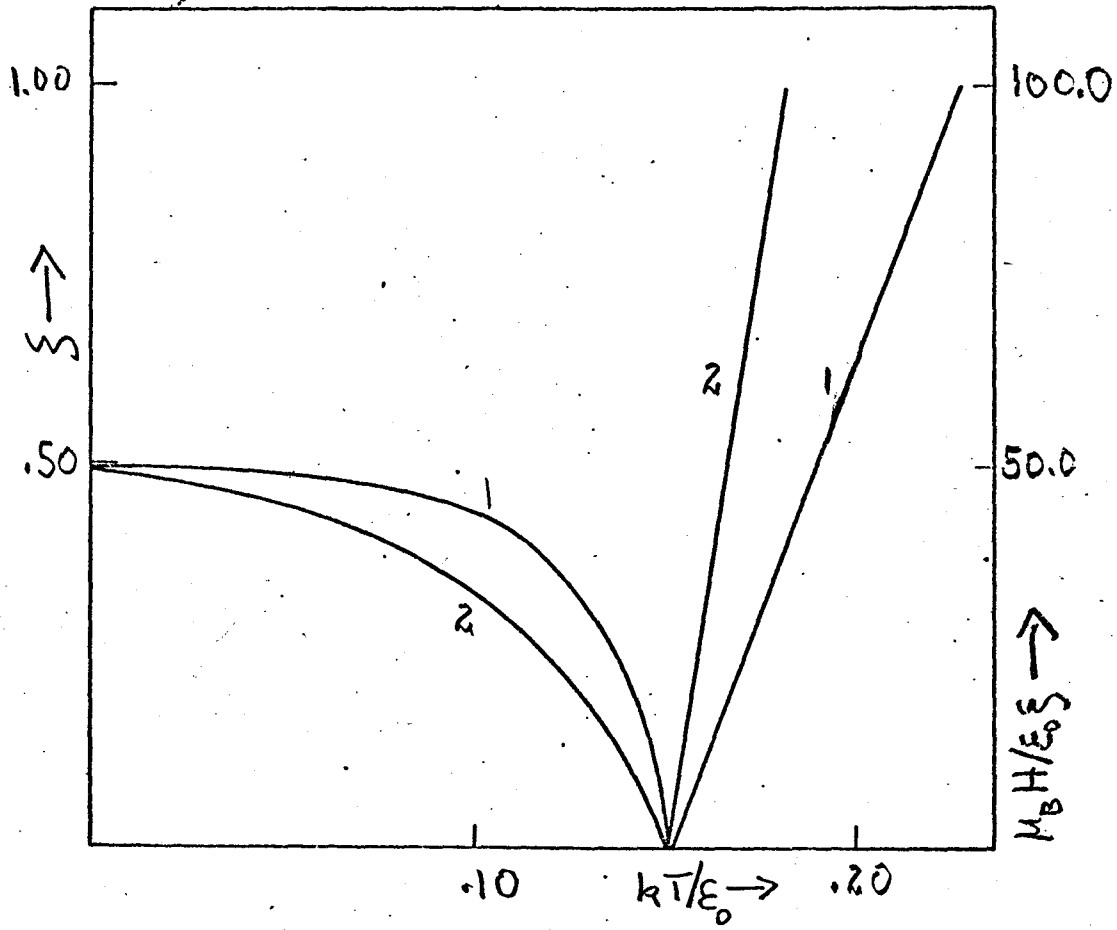


Fig. 3c

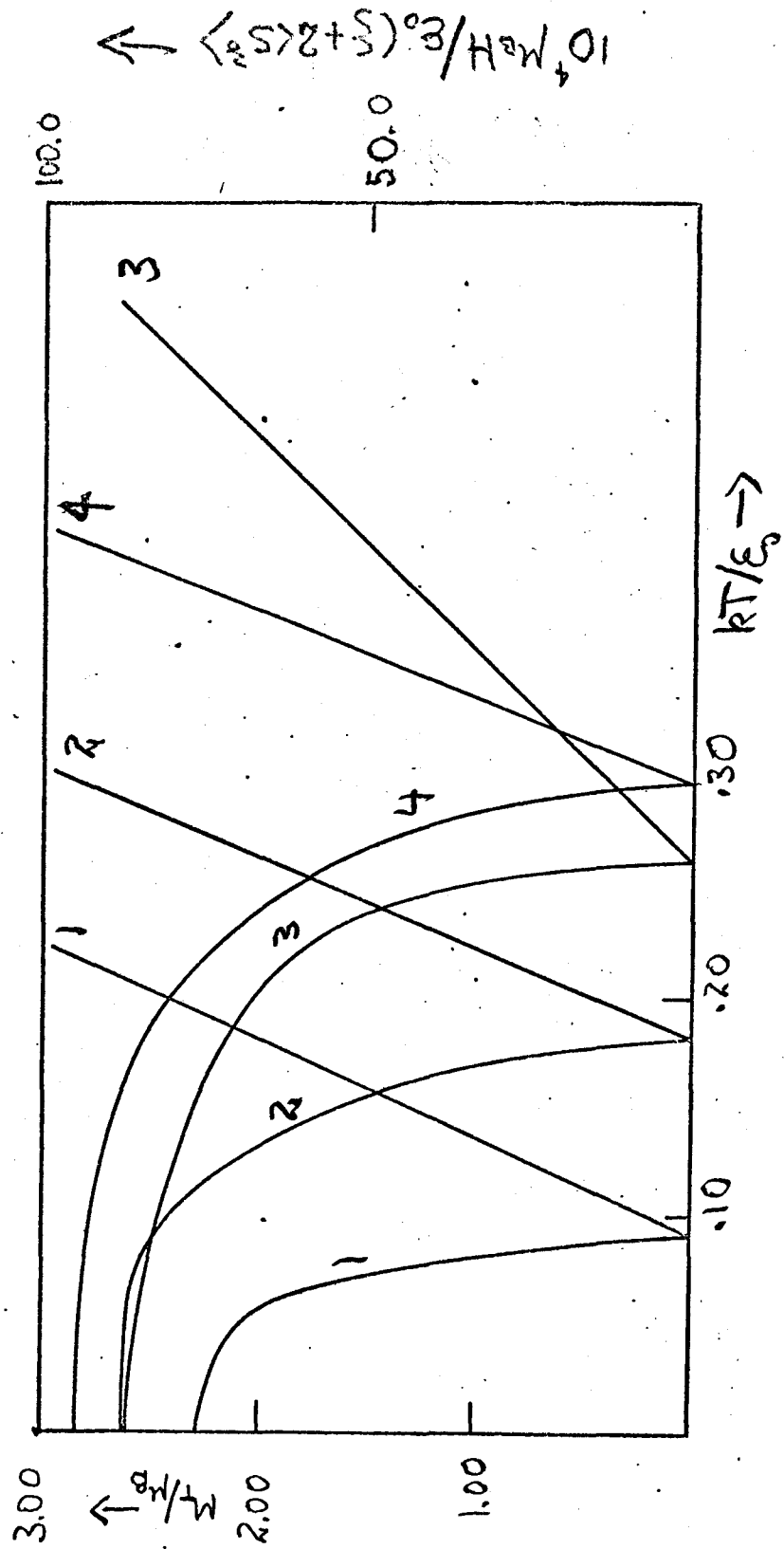


Fig. 3d